VACANCY TRANSPORT AND INTERACTIONS ON METAL SURFACES

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Vacancy Transport and Interactions on Metal Surfaces

Final Report

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Abstract

This proposal is a study of vacancy transport and vacancy interaction on metal surfaces. Adatom self-diffusion and vacancy diffusion on surfaces Au(100), Ag(110) and W(100) were subject of our investigation. The phenomena were investigated in ultra-high vacuum conditions with atomic scale resolution with Scanning Tunneling Microscope (STM) and Field Ion Microscope (FIM). For each investigated material, the research was divided into three stages: investigation of surface morphology, measurements of adatom surface self-diffusion, and measurements of vacancy diffusion. The key results and final status for each investigated surfaces are as follows:

The experiments conducted on Au(100)

The experiments of diffusion of Au adatom and surface vacancy on Au(100) surface were conducted with STM located Center for Microanalysis of Materials at University of Illinois at Urbana-Champaign. The main goal of the studies was to investigate diffusion behavior of single adatoms and single vacancies on Au(100) surface. Additional goal of the study was to investigate via diffusion measurements the interactions between vacancies to possibly form vacancy islands.

The morphology of Au(100) surface was determined as hex-Au(100) with <001> oriented one-dimensional channels. The investigation on adatom self-diffusion were conducted via measurements of island saturation on Au(100) surface. The activation energy E_A for adatom diffusion was determined 0.32±0.02eV. Obtained results with detailed description of measuring method were published in Phys. Rev. B **84**, 245445 (2011) and presented on 28th European Conference on Surface Science (ECOSS-28) by A. Trembulowicz, G.Ehrlich and G. Antczak.

In the second stage of the experiment, the experimental conditions for creation of surface vacancies on Au(100) has been determined and observations of surface diffusion of mono vacancies has been conducted. The vacancies were created via Ne^+ ion sputtering of Au(100) surface,the energy of bombarding ions was 100 – 150 eV and the time of exposure was 10 - 30 min, pressure of Ne gas was measured to be 10^{-7} Tr. The observations of vacancy diffusion delivered preliminary values of jump rates (Fig. 1).

At the time of this report, the value of activation energy for vacancy diffusion and the diffusivity pre-exponential factor has not been determined yet. However, supplementary measurements are planned with the results obtained to be published with acknowledgment of sponsorship from US Air Force.

The experiments conducted on Ag(110)

The experiments on diffusion of surface vacancies on Ag(110), were conducted with STM located Center for Microanalysis of Materials at University of Illinois at Urbana-Champaign. The main goal of the studies was to investigate diffusion behavior of single vacancies on Ag(110) surface. An additional goal of the study was to investigate the interactions between vacancies and their possibility to form vacancy islands. The obtained results were planned to be compared with results obtained for Au(100).

The experimental conditions necessary for cleaning Ag(110) surface has been determined. The preliminary parameters for vacancy creation in Ag(110) surface has been determined: $I_{sputtering} = $ 1-6 mA, $p_{Ne} = 9.8*10^{-7}$ Tr, $t_{sputtering} = 120$ s.

To investigate the surface diffusion of single adatoms and mono-vacancies, it was crucial to obtain the atomic resolution of Ag(110) surface. However the electrical noise which disturbed the information coming from the STM tip did not prevent obtaining systematical pictures with atomic scale resolution. Thus the experiments on adatom and mono-vacancy surface diffusion on Ag(110) were not been accomplished.

The experiments conducted on W(100)

The experiments on surface diffusion of W adatom and surface vacancy on W(100), were conducted with FIM located in Materials Research Laboratory at University of Illinois at Urbana-Champaign. The goal of the research was to determine the morphology of W(100) surface, find the value of activation energy for W adatom diffusion, determine the mechanism for adatom diffusion and obtain the activation energy for vacancy diffusion. Additional adsorption experiments of rhodium atoms on W(100) surface were conducted to confirm the morphology of W(100) surface.

There are three major competitive models of morphology of W(100) surface: non reconstructed, c(2x2)-reconstructed with lateral shift of surface's atoms (proposed by Debe and King the "zig-zag" model (Fig.) [1]) and c(2x2)-reconstructed with vertical shift of surface's atoms (proposed Tung, Graham and Melmed [2]). Conducted experiments on adsorption of Rh atoms and W adatom diffusion on W(100), showed incorrectness of Tung-Graham-Melmed model, which in spite of intense debate about this model, has never been shown before. The adsorption experiments did not show lateral reconstruction of W(100) surface which might be a result of the small plane size and the step edge relaxation [3]. For both models, it was previously reported [1] that the reconstruction disappears above the room temperature. The adatom diffusion experiments

were conducted at temperatures 66-726K. Thus the correctness of any of assumed models had no influence on adatom diffusion. However, considering that Tung-Graham-Melmed model of W(100) was a basis for proposed method for vacancy creation via field evaporation it was necessary for searching alternative methods of producing vacancies in W(100) surface.

The activation energy (E_A) for diffusion of W adatom has been fund 1.58±0.19 eV. Since recently collected data has not been yet included into our analysis, this value might slightly change after the process of data analysis is finished. Obtained value of activation energy for adatom diffusion differs from theoretical values proposed by Spisák and Hafner [4]: for the adatom exchange i.e. 2.3 eV, for adatom jump i.e.1.2 eV along the "zig-zag" and 1.3 eV perpendicular to the "zig-zag".

Based on analysis of adsorption sites of diffusing W adatom, the occurrence of the exchange mechanism on W(100) has been shown, which has never been done before for any of bcc(100) surfaces. In addition, it has been shown a first experimental proof and temperature dependence for occurrence of two diffusion mechanisms on the same surface: jump and exchange. Obtained results have yet to be published in journal article and in PhD dissertation of UIUC graduate student Tomasz Olewicz.

Concerning vacancy diffusion, pictures of surface mono-vacancies were resolved in FIM (Fig. 3). The surface vacancies were created by using two alternative methods: field evaporation and Ne^+ ion bombardment. The vacancies created by field evaporation were at the plane edge. Thus at higher temperatures due to decomposition of the W(100) surface, it was not possible to determine if the vacancy was merged into the step edge or if the step edge atoms detached first . Alternatively, the vacancies were created via Ne^+ ion sputtering. Used parameters of sputtering process are as follows: the sputtering current $I_{sputtering}$ =2*10^{-8} A, the time of Ne^+ ion bombardment $t_{sputtering}$=15s, the pressure of Ne gas p_{Ne}=1*10^{-4} Tr. The vacancies were created about 1-2 adsorption sites from the plane edge. However with introducing Ne gas into the system, significant amount of hydrogen and other residual gasses was introduced as well, which did not allowed for obtaining reliable data. A serious technical problem with the apparatus (i.e major crack of the glass FIM chamber and oxidation of all tungsten-based apparatus filaments) caused significant delay in the experiment's schedule and we were unable to investigate vacancy diffusion further before the end of the grant.

Personnel supported on the Project

The project supported two postdoctoral research associates and one graduate student, who performed the work under the supervision of the principal investigator. Dr. Artur Trembulowicz completed his degree from the University of Wroclaw in Poland where his focus was on the adsorption process and formation of ordered surface structures and alloys on metal substrates. He served as the lead postdoc on the project from October 2, 2009 through January 15, 2012 and was responsible for the supervision of daily activities in the lab and of the graduate student, and coordination with the PI on the planning and direction of the research. He left to return to his home country, where he accepted a position with the University of Wroclaw. He returned for approximately three (3) months as a visiting scholar starting in July of 2012 to work with the current postdoc and graduate student on the research. Dr. Marta Trelka was recruited and joined the research group in June of 2011 when Dr. Trembulowicz indicated his intention of returning home. She remained through the end of the project, taking on the tasks that Dr. Trembulowicz had performed when he left. She earned her Ph.D. from the Universidad Autonoma de Madrid in December 2010 and brought a background in fabrication and characterization of organic nanostructures by scanning tunneling microscopy to the work. Tomasz Olewicz earned his Master's Degree from the University of Wroclaw in experimental physics. He began as a visiting scholar in January of 2009 following the award the project. Upon his acceptance into the Materials Science and Engineering Ph.D. program at the University of Illinois, he served as the graduate research assistant for the project through its completion. Mr. Olewicz subsequently transferred to Electrical and Computer Engineering and plans to complete his PhD in August 2014. Mr. Olewicz was responsible for the daily experiments that produced the results of the research and wrote the final report for the project following Professor Ehrlich's unexpected death.

Publications resulting from research conducted on the Grant

1. Artur Trembułowicz, Gert Ehrlich, Grażyna Antczak,*Surface diffusion of gold on quasihexagonal-reconstructed Au(100)* ,Physical Review B **84** (2011) 245445-1-5.

2. *Coexistence of exchange and jump: W on W(100)*, submitted to Physical Review

3. Tomasz Olewicz PhD Dissertation, *Coexistence of two diffusion mechanisms: adatom jump and adatom exchange, for W on W(100)*, expected completion August 2014
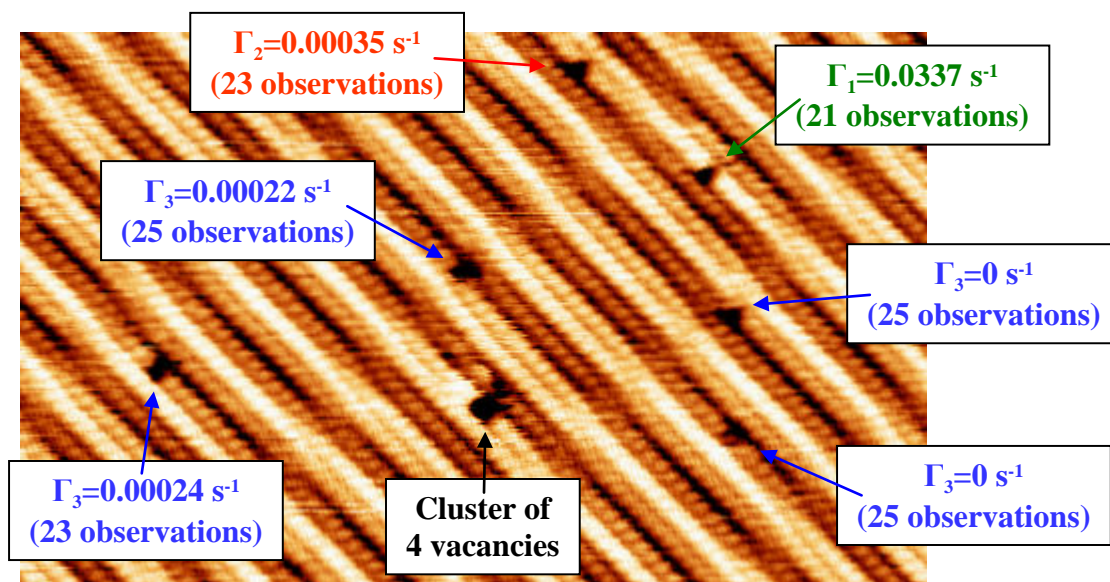
$\Gamma_2=0.00035$ s^{-1}
(23 observations)

$\Gamma_1=0.0337$ s^{-1}
(21 observations)

$\Gamma_3=0.00022$ s^{-1}
(25 observations)

$\Gamma_3=0$ s^{-1}
(25 observations)

$\Gamma_3=0.00024$ s^{-1}
(23 observations)

Cluster of
4 vacancies

$\Gamma_3=0$ s^{-1}
(25 observations)

Fig.1. Mono-vacancies and vacancy cluster on Au(100) surface with preliminary results of jump rates.
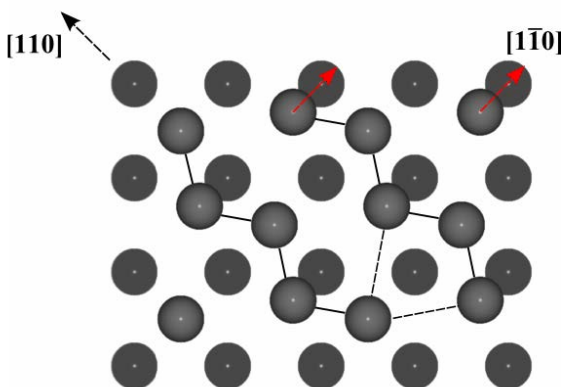

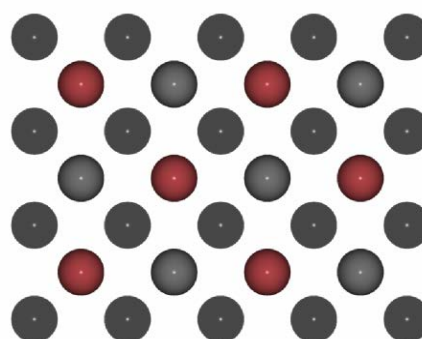Fig.2a. Debe-King model of W(100) surface.
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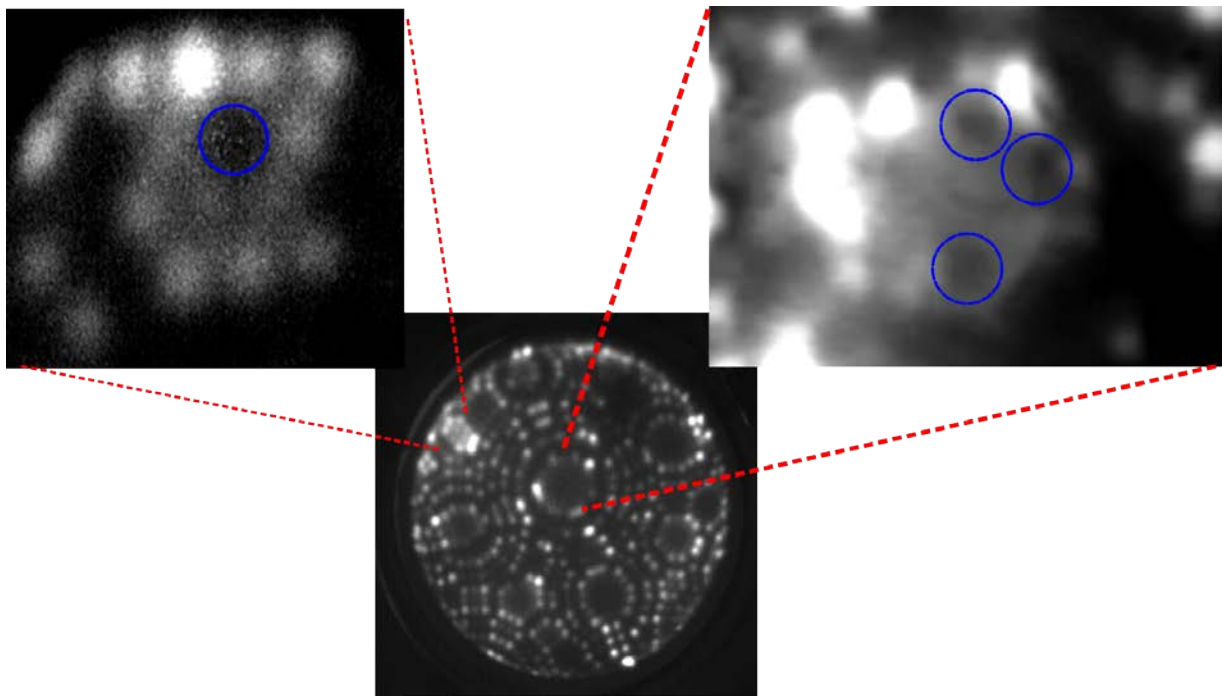Fig.2b Tung-Graham-Melmed model of W(100) surface.

Fig.3. FIM pictures of tungsten surface. Top left: picture of (211) plane with surface vacancy (blue circle), created by Ne^+ ion sputtering. Top right: picture of (100) plane with surface vacancies (blue circles) created by Ne^+ sputtering. Bottom center: picture of a whole W(100) oriented tip.

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Surface diffusion of gold on quasihexagonal-reconstructed Au(100)

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The scanning tunneling microscope has been used to measure the saturation island density N_x of gold on the hexreconstructed Au(100) surface over a range of temperatures starting at 76 K. Assuming that the critical island size equals one, $\partial \ln N_x / \partial(1/T) = \chi E_d/k$, where χ is the scaling exponent, E_d gives the activation energy for surface diffusion, and k is the Boltzmann constant. The scaling exponent χ has been obtained as 0.26 ± 0.03 from measurements of the island density as a function of the deposition rate, indicating that diffusion is indeed one-dimensional (anisotropic) and the critical island size is unity. We therefore derive an activation energy of 0.32 ± 0.02eV and a frequency prefactor of $2(\times 4^{\pm 1}) \times 10^{13}$ s^{-1} for diffusion of gold on hex-reconstructed Au(100).

I. INTRODUCTION

With the increasing importance of miniaturization in everyday life, the significance of understanding basic processes occurring at surfaces grows. One of the important events controlling the stability of nanostructures is surface diffusion. We have investigated diffusion on gold, a widely used material in technology. To our surprise, the basic process—surface diffusion on real reconstructed surfaces of Au(100)—is not well understood. The long-range reconstruction of the gold surface[1–10] establishes an environment not easy to investigate. The movement of gold atoms on the gold (100) plane is thus of scientific interest, but the kinetics of migration over this plane are still uncertain, and the complicated reconstruction is still a challenge to modeling.[10]

The only available diffusion measurements have been made by Günther et al.[11] with the scanning tunneling microscope (STM) to probe the saturation island density[12,13] N_x at different temperatures T, given by nucleation theory as

$$N_x \sim \theta^{\chi/i}(D/F)^{-\chi}\exp(\chi E_i/ikT). \quad (1)$$

Here θ is the surface coverage, D gives the surface diffusivity, F is the rate of deposition, and E_i is the binding energy of the critical cluster i; the binding energy is equal to zero for a critical cluster size of one. For two-dimensional diffusion $\chi = i/(i+2)$, whereas in one-dimensional (anisotropic) diffusion the exponent becomes $\chi = i/(2i+2)$.[13]

The first experiments on Au(100), carried out by Günther et al.,[11] found a scaling exponent $\chi = 0.37 \pm 0.03$, which initially appeared consistent with two-dimensional diffusion and a critical island size i of unity. Simulations, however, yielded values of the island density 7–10 times the experimentally determined density, and Günther et al., therefore, assumed one-dimensional (anisotropic) diffusion and approximated the experimentally observed saturation density using $E_d \sim 0.2$ eV with a critical island size of three and a trimer binding energy of 0.6 eV.

These findings were challenged by Liu et al.,[14] who considered the role of dimer diffusion, and with two-dimensional diffusion and a critical island size equal to one came up with an activation energy $E_d = 0.35$ eV for monomers and 0.45 eV for dimers, with a binding energy of 0.4 eV. Liu et al. assumed that reconstruction could not be so extensive that movement

changed from two- to one-dimensional, but they did not carry out control measurements and only reanalyzed the existing experimental data.

Subsequently the work of Günther et al.[11] was reanalyzed by Bartelt et al.[15] who showed that the results of Günther et al.[11] must be preferred. However, the island densities of Günther et al. were measured at elevated temperatures, from 315 to 435 K, a fairly large range, over which dimers may be mobile. The extensive reconstruction of the Au(100) surface may indeed influence the dimensionality of diffusion, and we therefore decided to resolve existing uncertainties by carrying out measurements over a lower temperature range than explored previously.

II. RESULTS

In order to explore clusters with a critical nucleus of one, we have repeated studies of the saturation island density of gold on Au(100) at low temperatures and at a pressure $<10^{-10}$ Torr. Measurements of the island density were done with an Omicron VT STM, always scanning at 100 K or lower, cooled with liquid helium to temperatures as low as 76 K; the temperature was measured by a thermocouple in contact with the top surface of a dummy sample. Values of the island density were determined separately a few times and were then averaged. An image of the clean gold surface, obtained by repeated ion bombardment at 1 keV for 15 min with argon, followed by 10 min annealing at 850 K, is shown in Fig. 1(a). The unit cell there, (6 × 30), comes about as the outermost layer is reconstructed into an arrangement resembling a quasihexagonal structure on top of a square lattice, illustrated in Fig. 2. The surface depicted in Fig. 1(a) is very similar to that of Pt(100)[16] and has channels half a unit cell long in the [01$\bar{1}$] direction. This is also demonstrated in Figs. 1(b) and 1(c) by the STM profiles along A and B across a unit cell. Reconstructed domains are observed with channels along [011] as well a [01$\bar{1}$].

When gold atoms are evaporated onto the Au(100) surface at ~200 K from a heated source, either a tungsten basket or a graphite crucible, stringlike islands are formed along the ⟨011⟩ rows, as is apparent in Fig. 3(a). Many of the same structures are seen at 236 K, in Fig. 3(b), but at a temperature of 120 or 90 K, in Figs. 3(c) and 3(d), the islands
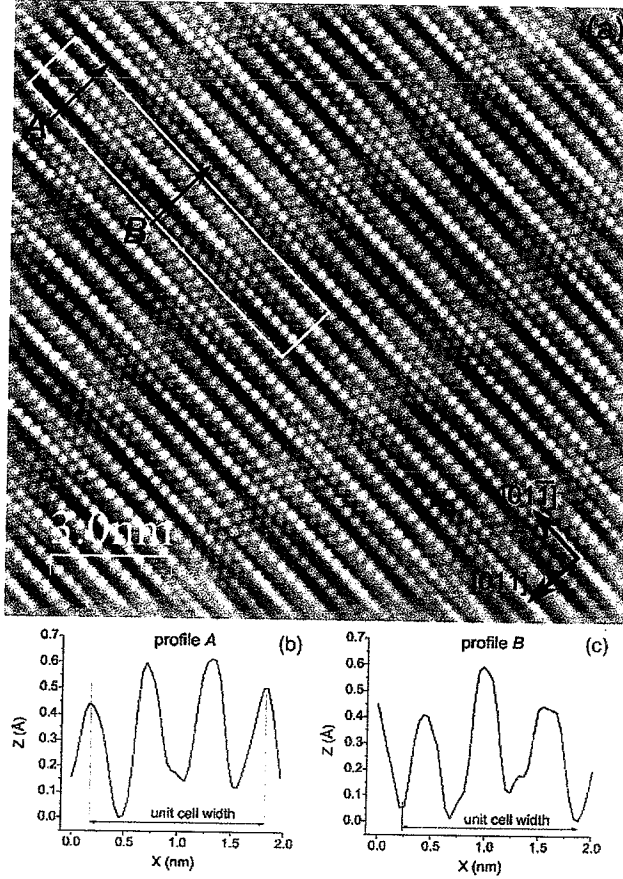
FIG. 1. (Color online) (a) STM image of reconstructed Au(100) surface at 3 nA and 2 mV, showing a (6×30) unit cell after bombardment at 1 keV with argon ions and annealing at 850 K. Scanning is along the horizontal. (b) Profile along A across a unit cell. (c) Profile along B.
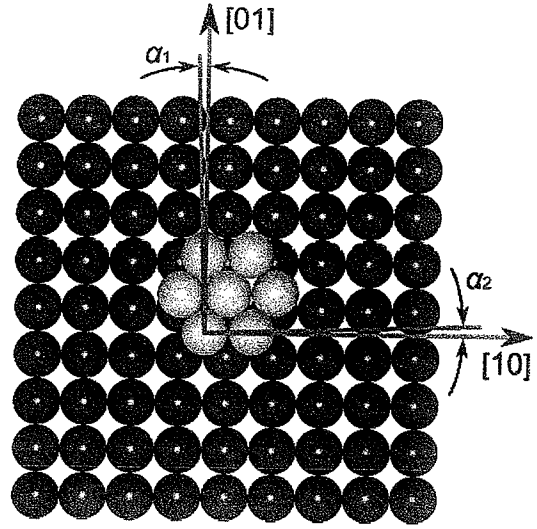

FIG. 2. (Color online) Model of quasihexagonal surface layer on Au(100) lattice (Ref. 6). $\alpha_1 = \alpha_2 \approx 0.7$ with respect to the [01] direction.

For the diffusivity expressed in terms of the jump length, the prefactor D_o is given by the essentially constant frequency prefactor ν. As is clear from Eq. (1), measurement of the saturation island density N_x at different temperatures, but at constant deposition rate F and constant coverage, should yield the activation energy E_d for diffusion, inasmuch as

$$\partial \ln N_x / \partial (1/T) = -\chi \partial \ln D / \partial (1/T) = \chi E_d / k. \quad (3)$$

are essentially round. Apparent in the shape of the islands is the slow diffusion rate with D/F below 10^5, known as the postnucleation regime. At 200 K, the aspect ratios of the islands vary but are as large as 1:37, which suggests preferred unidirectional (anisotropic) diffusion. Deposition is continued until the island layer is saturated, and the density of islands is obtained, as illustrated in Fig. 4 at 100, 150, and 200 K. As the temperature is increased from 100 K, the saturation island density diminishes but is still on the horizontal part of the curve of island density versus coverage. Saturation in general was reached by depositing the same amount of gold at other temperatures. An interval of ~75 min was necessary to reach saturation; thereafter, up to one-half hour was spent for the change from deposition to scanning temperature. A few hours were devoted to measurements. We assume that all single atoms attached to existing islands during the deposition, and there is no significant postdeposition activity during cooling to 100 K.

The diffusivity D can be written as a function of the temperature as

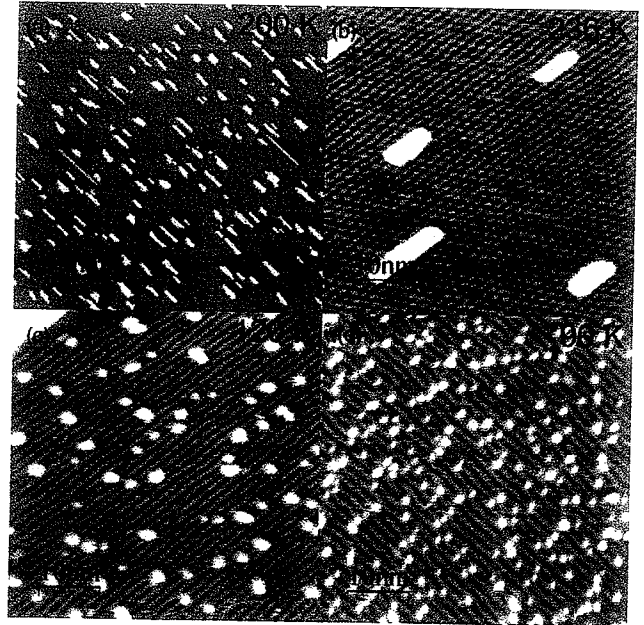$$D = D_0 \exp(-E_d/kT). \quad (2)$$

FIG. 3. (Color online) STM image of gold islands deposited on reconstructed Au(100). (a) For Au(100) temperature of 200 K, islands are stringlike. Scanning is at 100 K with 0.5 nA and 1 V. (b) At T = 236 K islands are stringlike. (c) At T = 120 K and (d) at T = 90 K, islands are round.
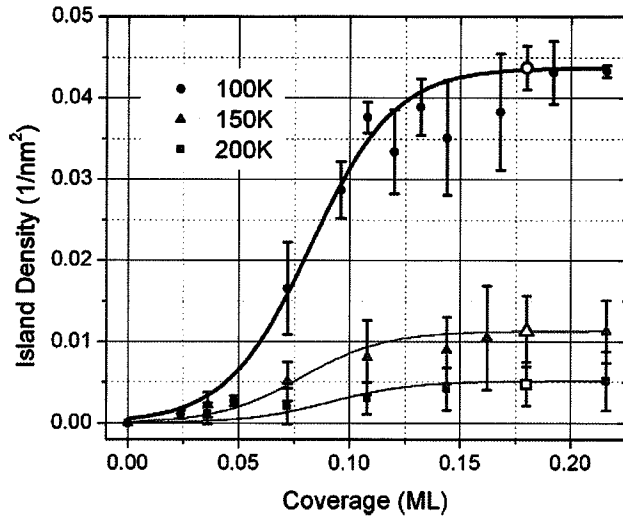
FIG. 4. (Color online) Plot of island density versus coverage of gold on reconstructed Au(100) at $T = 100$, 150, and 200 K. Open symbols indicate saturation density.

This, of course, assumes that the critical island size is equal to one. A plot of the logarithm of the saturation island density N_x against the reciprocal temperature $1/T$ over the range from 76 to 234 K is given in Fig. 5, with the line drawn only as a guide. Scanning was always done at 100 K. On the assumption that the critical cluster size is unity and diffusion is highly anisotropic (one dimensional), the slope of the curve from 156 to 236 K yields an activation energy of 0.32 ± 0.02 eV.

The important question now is the size of the critical cluster: is it indeed unity? To explore this matter, we have in addition measured the island density N_x as a function of
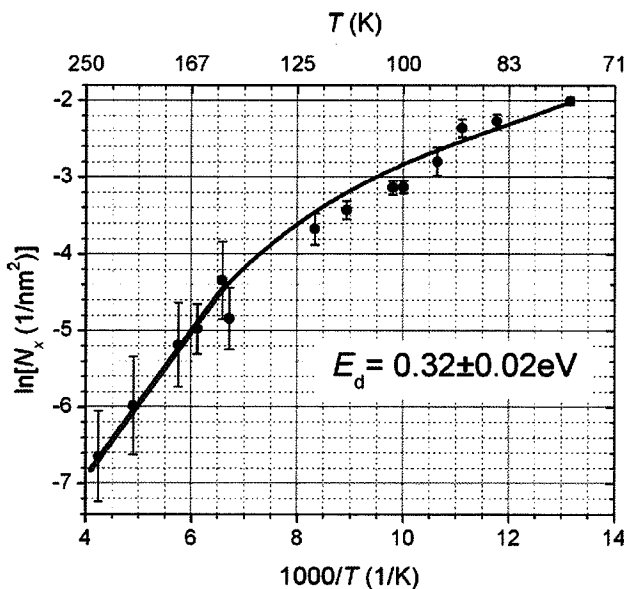

FIG. 5. (Color online) Plot of logarithm of the gold saturation island density on reconstructed Au(100) as a function of $1/T$. Analysis assumes one-dimensional (anisotropic) diffusion and a critical island size of one. E_d derived for data from 236 to 156 K, shown by heavy (red) line. Curve is for guidance only.
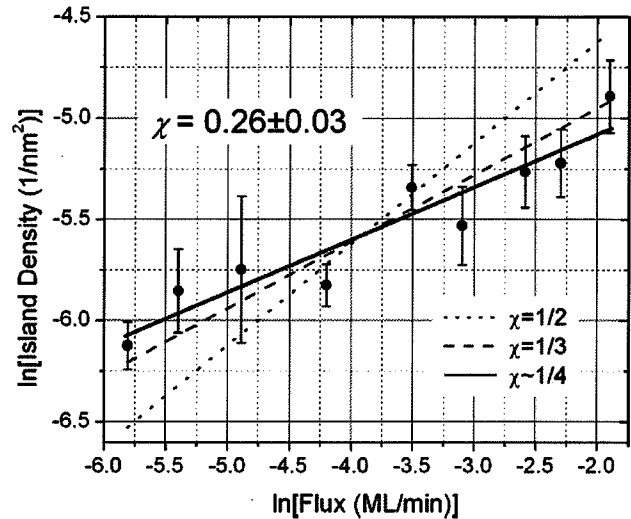

FIG. 6. (Color online) Plot of saturation island density as a function of the deposition rate F on surface at 200 K, yielding a scaling exponent χ equal to 0.26 ± 0.03.

the deposition rate F on the Au(100) surface. The rate F has been calibrated by determining the Rutherford backscattering of gold deposited on silicon. By varying the flux by a factor of $\sim$50, we have, as illustrated in Fig. 6, found that $\chi = 0.26 \pm 0.03$. The value of the exponent χ is that expected for highly anisotropic (one-dimensional) diffusion and a critical cluster size of unity, as assumed in our analysis.

That diffusion is unidirectional seems to be confirmed by Fig. 7, which was taken at $T = 100$ K under manipulation conditions, after neon sputtering at $\sim$150 eV for 20 min. After sputtering, gold atoms are present on the surface. Scanning the surface horizontally, starting at the bottom of the image, reveals the path of a single adatom propelled over the surface by the tip. The adatom moves over a considerable distance along one [01$\bar{1}$] atom row until, as indicated by arrows, it jumps to an adjacent one, where it continues its motion, which demonstrates that diffusion in the $\langle 011 \rangle$ direction is preferred. This strong preference in the direction indicates an easy path for adatom diffusion. We believe that this preference is caused by the structure of the surface. In a subsequent image the adatom does not appear, most likely having become attached to the scanning tip and being removed.

III. DISCUSSION

Our result of 0.32 eV for the activation energy of self-diffusion is in good agreement with the value of 0.35 eV reported by Liu *et al.*[14] on the presumption of two-dimensional motion and a critical island size equal to one. Our value for the activation energy was, however, derived for anisotropic motion, as was the barrier of $\sim$0.2 eV of Günther *et al.*[11] obtained at a much higher temperature $T \geqslant 315$ K. Under these conditions it is likely that gold dimers will be mobile and may influence the saturation island density. At the much lower temperatures explored by us, only single atoms are likely to diffuse.
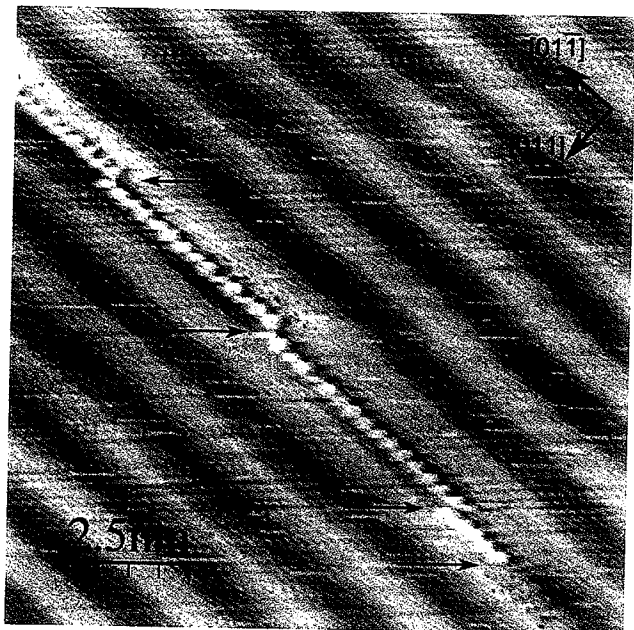
FIG. 7. (Color online) STM image of a gold adatom, produced by neon sputtering, moved over the reconstructed Au(100) by the scanning tip. Scanning at 100 K under manipulation conditions, at 20 nA and 12 mV, is along the horizontal, from the bottom to the top. The adatom moves over an appreciable length along one [01$\bar{1}$] atom row, until as indicated by arrows, it jumps to an adjacent one, where it continues its movement. A subsequent STM image does not show the atom.

Because we know the deposition rate to be 0.0025 ML/min (with 1 ML = 1×10^{15} atoms/cm^2) and the coverage θ is 0.18, we find from the intercept of the diffusivity at infinite temperature, a frequency prefactor of $2(\times 4^{\pm 1}) \times 10^{13}$ s^{-1}. We can now also raise the obvious question—why did we only analyze the curve of the island density above 156 K? The answer is that the ratio of D/F amounts to 5×10^6 at $T = 150$ K but is only 3×10^4 at 125 K, below the limit of 10^5 for obtaining a reasonable diffusivity; at these low temperatures the island density is no longer adequately given by simple nucleation theory,[12] so these measurements have to be disregarded.

A surface structure similar to that of hex-Au(100) is that of hex-Pt(100),[16] and Linderoth et al.[17] have used an STM to examine the self-diffusion of platinum on this surface. For anisotropic diffusion and a critical island size of one, they arrived at an activation energy of 0.43 eV for surface diffusion. The ratio of the activation energy to the heat of sublimation is 0.0814, compared to 0.0943 for our results for hex-Au(100), a ratio typical for surface self-diffusion on similarly structured surfaces.[18] In contrast, the diffusion energy of Günther et al.,[11] ~0.2 eV, gives a much lower ratio of only 0.058, which does not correspond with the value for hex-Pt(100).

For nonreconstructed Pt(100), diffusion has been observed to occur by exchange with the substrate,[19] which is also the mechanism indicated in several theoretical studies[20–29] for both Pt(100) and Au(100). It is, however, not clear that on the reconstructed surface exchange is a significant mechanism. It may well be that the compression of the reconstructed surface is able to eliminate the exchange mechanism from the picture.[28] For hex-Au(100) as well as hex-Pt(100), there are no experimental indications available for the mechanism of diffusion, and we must emphasize that there is no information about the diffusion mechanism emerging from our studies.

IV. CONCLUSIONS

We have demonstrated that diffusion of gold over the hex-reconstructed Au(100) surface is one dimensional and proceeds primarily along the ⟨011⟩ channels created during reconstruction. The exchange process suggested for unreconstructed Au(100) is most likely suppressed by the compression of the surface layer, but no direct information is available about the diffusion mechanism on the reconstructed Au(100) surface. However, we have identified an effective barrier of 0.32 ± 0.02 eV and a frequency prefactor of $2(\times 4^{\pm 1}) \times 10^{13}$ s^{-1} to unidirectional surface self-diffusion.

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